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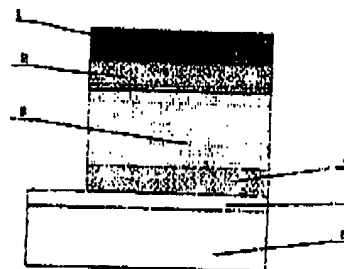
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[54] 实用新型名称 高效高色纯度镧系金属配合物有机电致发光器件

[57] 摘要

本实用新型属于电致发光技术领域,是一种高效高色纯度长寿命镧系金属配合物有机电致发光器件。主要由电子传输层、混合层、空穴传输层和 ITO 导电膜等构成,其中在电子传输层和空穴传输层之间的混合层是两者的过渡层。混合层是由三价镧系金属离子配合物 (InC 表示,其中 L 为镧系金属, C 配体) 和空穴传输层材料混合而成,电子传输层材料可以是 LC 也可以是不发光镧系金属与相同配体 C 的配合物。采用这种器件结构既可以防止与空穴传输层生成基态复合物,保证镧系金属离子的高色纯度带发射,又可以提高器件的量子效率。



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权 利 要 求 书

1、一种高效高色纯度镧系金属配合物有机电致发光器件，其结构包括金属电极（1），电子传输层（2），空穴传输层（4），ITO导电膜（5），透明玻璃基板（6），其特征是发射层为混合层（8），混合层（8）在空穴传输层与电子传输层之间，混合层材料由三价镧系金属离子配合物和空穴传输层材料混合而成，从空穴传输层到电子传输层，混合比例逐渐递减，电子传输层材料可以是三价镧系金属离子配合物InC，也可以是不发光L镧系金属与C的配合物。

2、根据权利要求1所述的高效高色纯度长寿命镧系金属配合物有机电致发光器件，其特征是混合层（8）中三价镧系金属离子配合物与空穴传输层材料的混合比例从空穴传输层由HTL:InC=6:1~3:1逐渐变化到HTL:LnC-1:6~1:3。



说明书

高效高色纯度镧系金属配合物有机电致发光器件

本实用新型属于电致发光技术领域，是一种镧系配合物有机电致发光器件。

有机电致发光器件由有机小分子材料以薄膜叠加而成，其中发光材料必须是镧系配合物发光体。具体器件结构是：正极/ITO/有机多层薄膜/阴极。正极为玻璃基板，阴极为金属电极，ITO为透明导电膜，是 In_2O_3 和 Sn_2O_3 混合的氧化物薄膜。在直流电压(5-15V)驱动下，可从透明阳极(正极)侧发出可见或红外荧光。

传统的有机电致发光(简称EL)器件结构均是前述的“三明治”式多层结构器件，这种结构的器件一般由三个有机功能层构成即：ITO/HTL(空穴传输层)/EML(发射层)/ETL(电子传输层)/M(金属)，如果EML兼有ETL或HTL功能时，则器件为两层结构即为ITO/HTL/ETL-EML/M，或者ITO/EML-HTL/ETL/M结构。这两类器件结构都存在明显的界面效应，特别是选用镧系金属配合物作为EML时，层间界面使驱动电压升高，在直流驱动下，发射光谱不是单纯由三价镧系离子的窄带构成，发射出来自EMI与二胺衍生物HTL材料(如TPD或NPB)形成基激复合物，使得电致发光(EL)发射波长明显移向长波，并且谱带变宽，严重干扰三价镧系金属离子的窄带发射。同时，还会使发光效率和驱动寿命明显降低。

为了防止由镧系配合物做发射层的有机电致发光器件出现基激复合物Exciplex宽带发射，获得纯的稀土窄带发射，和高色纯度的有机电致发光器件，提高用镧系配合物做发射层的有机电致发光器件的量子效率，进而提高器件的发光亮度，和提高用稀土配合物做发射层的有机电致发光器件的工作稳定性和工作寿命，本实用新型提供一种消除界面效应的高效、高色纯度长寿命的镧系金属配合物有机电致发光器件。

本实用新型由金属电极，电子传输层，混合层，空穴传输层，ITO导电膜，透明基板如玻璃等构成。金属电极为阴极，所用的金属为Mg:Ag。透明基板为阳极。混合层为发射层，是三价镧系金属离子配合物光发射材料和空穴传输层的混合体。混合层中的空穴传输层材料从空穴传输层到电子传输层的含量逐渐减少。在器件的结构中，既看不出空穴传输层与混合层之间有明显界面，也看不出电子传输层与混合层间的明显界面，即从空穴传输层向电子传输层过渡是个渐变过程，也就是说混合层实际上是一个过渡层。

附图是本实用新型的结构图。图中1为金属电极, 2电子传输层, 3混合层, 4空穴传输层, 5 ITO导电膜, 6透明基板。

本实用新型所用的是三价镧系金属离子配合物(以下用LnC代替), 它的通式是: $\text{Ln}^{3+}\text{L}_1\text{L}_2$, $\text{Ln}=\text{Pr}, \text{Nd}, \text{Sm}, \text{Eu}, \text{Tb}, \text{Ho}, \text{Er}, \text{Tm}, \text{Yb}$, 中的任何一种或几种金属的组合, L_1 =有机脂肪或芳香羧酸根, β -二酮等, L_2 =中性有机配体化合物, 如: 含任何取代基的联吡啶(R-phen), 2, 2'-联吡啶(bpy), 氧化三苯基胺(TOPO), 三正辛基氧化膦(TPPO)等。HTL是通常使用的二胺衍生物, 如TPD或NPB。具体工艺过程是先在事先洗净的ITO电极上镀上一层厚度为200-500nm的空穴传输材料层, 然后同时蒸发LnC材料使它与HTL的混合比例由HTL:LnC=6:1~3:1, 逐渐变化到HTL:LnC=1:6~1:3, 这就是混合层。再在混合层之上蒸镀一层厚度为200-500nm的ETL材料, 如果该LnC具有电子传输性能, ETL可以是该LnC, 如果LnC不具有电子传输性能, 那么, ETL可以选择LC材料, 其配体与LnC配体相同, L(镧系)金属离子为不发光三价离子 Y^{3+} , La^{3+} , Gd^{3+} 或 Lu^{3+} , 在最后形成的器件结构中, 既看不出HTL与混合层之间的明显界面, 也看不出混合层与ETL间的明显界面, 即从HTL层向ETL层过渡是个渐变过程。

本实用新型这种模糊的功能层间“界面”可防止混合层与HTL形成基激复合物发射, 可以获得纯的三价镧系离子的窄带发射。由于层间界面几乎不存在, 可以降低载流子注入势垒, 由于与HTL间的模糊界面, 避免基激复合物发射, 因此可以提高发光量子效率。因为在电激发下, 基激复合物发光波长明显向长波移动, 因而难于获得纯稀土窄带发射, 导致色纯度降低。需要说明的是这里所说的“混合式”与“掺杂式”结构不同, 前者混合比例大, 后者比例很小, 发光材料只占被掺杂材料的百分之几。选用空穴传输层为二胺, 如TPD或NPB, 器件结构为ITO/HTL/HTL:LnC/InC或LC/Mg:Ag, LNC为下表配合物时的主要结果:

序号	镧系配合物 (LnC)	配体化合物		发光效率 (%)	EL发射	
		第一配体	第二配体		主峰(nm)	发光
01	Eu(DBM)3phen	DBM:二苯甲酰甲烷	phen:邻二氮菲	2-3	611	红
02	Tb(ACAC)3phen	ACAC:乙酰丙酮	phen:邻二氮菲	1-3	544	绿
03	Nd(DBM)3phen	DBM:二苯甲酰甲烷	Bpy:2,2'-联吡啶		800,1000	红
04	Tb(ACAC)3phen	ACAC:乙酰丙酮	phen:邻二氮菲	0.9-1.0	480	蓝
05	Dy(ACAC)3phen	ACAC:乙酰丙酮	Bpy:2,2'-联吡啶	0.9-2.0	480,580	白

本实用新型的制作过程如下:

在真空度 10^{-4} 帕,所有有机材料热蒸发到ITO玻璃基板上,HTL厚度为30nm,混合层厚度50nm,ETL厚度为20nm,所有器件均在干燥空气中用紫外粘合剂把玻璃板压在器件上面封装成为有机EL(电致发光)器件。

实施例1

混合层材料是Tb(ACAC)₃phen:TPD由1:3变到3:1,ETL材料是Tb(ACAC)₃phen.这种器件在3V直流驱动下可发出Tb³⁺的纯线状光谱(主峰:545nm)。在低于 $200\mu\text{A}/\text{cm}^2$ 电流密度下量子效率可达3%以上。

实施例2

混合层材料是Eu(DBM)₃phen(三价铕与二苯甲酰基甲烷和喹啉的配合物)HTL材料是TPD,混合层比例Eu(DBM)₃phen:TPD由1:3变到3:1,这种器件在2.5V直流驱动下可发出Eu³⁺的纯线状光谱(主峰:611nm)。在低于 $200\mu\text{A}/\text{cm}^2$ 电流密度下量子效率可达4%以上。

实施例3

混合层材料是Dy(ACAC)₃phen,空穴传输层材料是TPD,混合层比例由1:3变到3:1电子传输层材料是Dy(ACAC)₃phen(三价铈与乙酰丙酮和喹啉的配合物)。这种器件在3V直流驱动下可发出Dy³⁺的纯线状光谱(主峰:480nm和580nm)。在低于 $200\mu\text{A}/\text{cm}^2$ 电流密度下量子效率可达1%以上。

实施例4

混合层材料是Er(DBM)₃phen(三价铈与二苯甲酰基甲烷和喹啉的配合物),空穴传输层材料是TPD,混合层比例Er(DBM)₃phen:TPD由1:3变到3:1.这种器件在2.5V直流驱动下可发出Er³⁺的纯线状光谱(主峰:525nm)。在低于 $200\mu\text{A}/\text{cm}^2$ 电流密度下量子效率可达1%以上。

实施例5

混合层材料是Tb(ACAC)₃phen,空穴传输层材料是TPD,混合层比例Tb(ACAC)₃phen:TPD由1:3变到3:1,材料是Gd(ACAC)₃phen(三价钆与乙酰丙酮和喹啉的配合物)。这种器件在3V直流驱动下可发出Tb³⁺的纯线状光谱(主峰:545nm),在低于 $200\mu\text{A}/\text{cm}^2$ 电流密度下量子效率可达3%以上。

实施例6

混合层材料是Tm(ACAC)₃phen,空穴传输层材料是TPD,混合层比例Tm(ACAC)₃phen:TPD由1:3变到3:1,电子传输层材料是Gd(ACAC)₃phen(三价钆与乙酰丙酮和喹啉的配合物)。这种器件在3V直流驱动下可发出Tm³⁺的纯线状光谱(主峰:480nm)。在低于 $200\mu\text{A}/\text{cm}^2$ 电流密度下量子效率可达3%以上。

实施例7

混合层材料是Nd(DBM)₃phen,空穴传输层材料是TPD,混合层比例Nd(DBM)₃phen:TPD由1:3变到3:1,电子传输层材料是Nd(DBM)₃phen.这种器件在6V直流驱动下可发出Nd³⁺的纯线状光谱(主峰:900nm和980nm),发红外光。

99-12-03

说明书附图

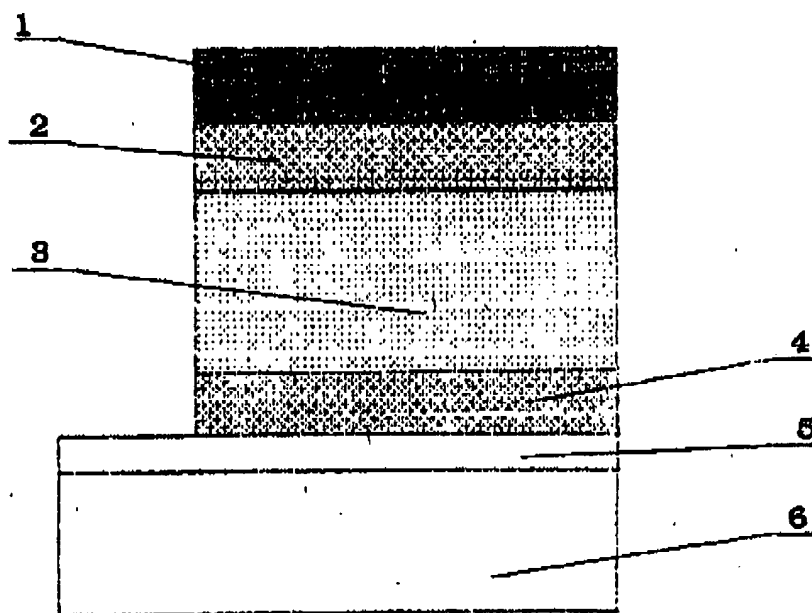


图 1

OA阶段特殊要求

(没有包含案卷和客户的一般性要求)

CPEL0252081P

2005年4月22日

【委托人OA特殊要求】

2004年6月份访问Philips后提出的具体要求：明确要求代理人要对审查意见进行分析
和评述，并就Philips公司的律师如何准备能够克服审查意见的答辩意见以及以后如
何起草、修改权利要求和 / 或说明书给出具体的指导性或建设性意见。

CN2421793Y

Title of utility model: Organic Electroluminescent Device of Lanthanide series Metal Complex with High Efficiency and High Color Purity

Abstract

This utility model relates to the technical field of electroluminescent, and is an organic electroluminescent device of lanthanide series metal complex with high efficiency, high color purity and long life span. Said device is mainly composed of electron transporting layer, mixed layer, hole transporting layer and ITO conductive film, etc., wherein the mixed layer disposed between the electron transporting layer and the hole transporting layer is the transition layer of these two layers. The mixed layer is formed of a mixture of the trivalent lanthanide series metal ion complex (represented by LnC , wherein L is the lanthanide series metal and C is the ligand.) and the hole transporting layer material, and the material of the electron transporting layer could be either LC or the complex of non-luminous lanthanide series metal and the same ligand C . Such device structure could both prevent generating the exciplex with the hole transporting layer, so as to ensure the high color purity band emission of the lanthanide series metal ion, and improve the quantum efficiency of the device.

Organic Electroluminescent Device of Lanthanide series Metal Complex with High Efficiency and High Color Purity

This utility model relates to the electroluminescent technical field, and is an organic electroluminescent device of lanthanide series metal complex.

Organic electroluminescent device is laminated by organic small molecule materials in the form of thin film, wherein the luminescent material must be the lanthanide series complex illuminant. The specific structure of the device is anode/ITO/organic multi-layer thin film/cathode. The anode is glass substrate, the cathode is metal electrode, ITO is transparent conductive film which is an oxide thin film mixed by In_2O_3 and Sn_2O_3 . When driven by DC voltage (5-15V), visible or infrared fluorescence could be emitted from the transparent anode (positive) side.

4 The structures of conventional organic electroluminescent (referred to EL) devices are all the aforementioned "sandwich" structure or multi-layer structure devices, which devices with this structure are generally composed of three organic functional layers, i.e., ITO/HTL(hole transporting layer)/EML (emission layer)/ETL (electron transporting layer)/M (metal). If the EML has the functions of both ETL or HTL, the device is a two-layered structure, i.e., ITO/HTL/ETL-EML/M, or ITO/EML-HTL/ETL/M structure. Both of said two kinds of device structures have obvious interface effect, especially when lanthanide series metal complex is selected to be EML, the interface between layers causes the driving voltage to increase, and under the DC driving, the emission spectrum is not merely consisted of the narrow band of trivalent lanthanide series ion, then it is emitted from EML and diamine derivative HTL material (e.g., TPD or NPB) to form the exciplex, thus the emission wavelength of electroluminescent (EL) moves to the long wave distinctly, and the spectrum band becomes wider, so that the narrow band emission

of trivalent lanthanide series metal ion will be seriously disturbed. Meanwhile, the luminous efficiency and driving life-span will be reduced distinctly.

In order to prevent the organic electroluminescent devices with lanthanide series complex as the emission layer from occurring exciplex broad band emission, and to obtain pure rare earth narrow band emission and high color purity organic electroluminescent device, and to improve the quantum efficiency of the organic electroluminescent devices using the lanthanide series complex as emission layer so as to improve the luminous brightness of the device, and to improve the working stability and life-span of the organic electroluminescent devices using the rare earth complex as the emission layer, the present utility model provides an organic electroluminescent device of lanthanide series metal complex with high efficiency, high color purity and long life-span, which could eliminate the interface effect.

The present utility model is composed of metal electrode, electron transporting layer, mixed layer, hole transporting layer, ITO conductive film, transparent substrate such as glass, etc. The metal electrode is the cathode and the metal used is Mg:Ag. The transparent substrate is the anode. The mixed layer is the emission layer, which is a mixture of light-emitting material of trivalent lanthanide series metal ion complex and the hole transporting layer. The hole transporting layer material in the mixed layer decreases gradually in content from the hole transporting layer to the electron transporting layer. In the structure of the device, neither obvious interface could be seen between the hole transporting layer and the mixed layer, nor any obvious interface could be seen between the electron transporting layer and the mixed layer, that is, the transition from the hole transporting layer to the electron transporting layer is a process of gradual change, and the mixed layer is actually a transition layer.

The figure is a structural schematic of the present utility model. In fig. 1, 1 represents the metal electrode, 2 represents the electron transporting layer, 3 represents the mixed layer, 4 represents the hole transporting layer, 5 represents the ITO conductive film and 6 represents the transparent substrate.

The present utility model uses the trivalent lanthanide series metal ion complex (hereinafter referred to LnC), whose general expression is a combination of one or several of the metals: $\text{Ln}^{3+}\text{L}_1\text{L}_2$, $\text{Ln}=\text{Pr, Nd, Sm, Eu, Tb, Ho, Er, Tm, Vb}$, wherein L_1 =organic fat or aromatic carboxylic acid, β -diketone, etc., L_2 =neutral organic ligand compound, such as R-phen with any substituent, 2, 2'-bpy, trioctylphosphine oxide(TOPO), triphenylphosphine oxide(TPPO), etc. HTL is the generally used diamine derivative, such as TPD or NPB. The specific processes are first coating a layer of hole transmission material with the thickness of 200-500nm on the ITO electrode that is cleaned in advance; then simultaneously evaporating LnC material to make the mix proportion with HTL to change from $\text{HTL}:\text{LnC}=6:1\sim 3:1$ to $\text{HTL}:\text{LnC}=1:6\sim 1:3$ gradually, and this is the mixed layer; next, evaporating a layer of ETL material with the thickness of 200-500nm on the mixed layer. If said LnC has the electron transporting property, ETL could be said LnC; and if LnC does not have the electron transporting property, ETL could select LC material, whose ligand is same as LnC ligand, and L (lanthanide series) metal ion is non-luminous trivalent ion Y^{3+} , La^{3+} , Gd^{3+} or Lu^{3+} . In the resultant device structure, neither any obvious interface between HTL and the mixed layer could be seen nor any obvious interface between the mixed layer and the ETL could be seen, that is, the transition from the HTL layer to ETL layer is a gradually changing process.

Such vague "interface" between functional layers in the present utility

model could prevent the mixed layer and the HTL from forming the exciplex emission, and could obtain narrow band emission of pure trivalent lanthanide series ion. Since the interface between layers nearly does not exist, the carrier injection barrier could be reduced, and owing to the vague interface with the HTL, the exciplex emission is avoided, thus luminous quantum emission efficiency could be improved. Under electrical excitation, the wavelength of exciplex emission moves to the long wave obviously, so it is hard to obtain pure rare earth narrow band emission and thereby resulting in decrease of color purity. It is necessary to explain that the "mixing type" structure mentioned herein is different from the "doping type" in structure, since the former has a large mix proportion while the latter has a very small mix proportion, and the emission material only takes a few percent of the doped material. The hole transporting layer is selected as diamine, such as TPD or NPB, and the device structure is ITO/HTL/HTL:LnC/InC or LC/Mg:Ag, and Lnc is the complex as indicated in the following table, then the main results are as shown in the following table:

No.	Lanthanide series Complex (LnC)	Ligand compound		Luminous efficiency (%)	EL emission	
		First ligand	Second ligand		Main Peak (nm)	emission
01	Eu(DBM) ₃ phen	DBM	phen	2-3	611	Red
02	Tb(ACAC) ₃ phen	ACAC	phen	1-3	544	Green
03	Nd(DBM) ₃ phen	DBM	Bpy:2, 2'-bpy		900,1000	Red
04	Tm(ACAC) ₃ phen	ACAC	Phen	0.9-10.	480	Blue
05	Dy(ACAC) ₃ phen	ACAC	Bpy:2, 2'-bpy	0.9-2.0	480,580	White

The manufacturing process of the present utility model is as follows:

Under the vacuum degree of 10^{-4} Pa, all the organic materials are evaporated onto the ITO glass substrate with heat, and the thickness of HTL is 30nm, the thickness of the mixed layer is 50nm, the thickness of ETL is 20nm, and all the devices are packaged into organic EL (electroluminescent) devices by pressing the glass plate onto the devices with ultraviolet adhesive in dry air.

Embodiment 1

The material of the mixed layer is $\text{Tb}(\text{ACAC})_3\text{phen}:\text{TPD}$ which changes from 1:3 to 3:1, and the material of ETL is $\text{Tb}(\text{ACAC})_3\text{phen}$. Such device could emit pure linear spectrum (main peak: 545nm) of Tb^{3+} under the DC drive of 3V. The quantum efficiency could reach above 3% under the electric current density lower than $200 \mu \text{A}/\text{cm}^2$.

Embodiment 2

The material of the mixed layer is $\text{Eu}(\text{DBM})_3\text{phen}$ (the complex of trivalent europium and DBM and phen), the material of HTL is TPD, and the proportion of the mixed layer $\text{Eu}(\text{DBM})_3\text{phen}:\text{TPD}$ changes from 1:3 to 3:1. Such device could emit pure linear spectrum (main peak: 611nm) of Eu^{3+} under the DC drive of 2.5V. The quantum efficiency could reach above 4% under the electric current density lower than $200 \mu \text{A}/\text{cm}^2$.

Embodiment 3

The material of the mixed layer is $\text{Dy}(\text{ACAC})_3\text{phen}$, and the material of the hole transporting layer is TPD, the proportion of the mixed layer changes from 1:3 to 3:1, and the material of the electron transporting layer is $\text{Dy}(\text{ACAC})_3\text{phen}$ (the complex of trivalent dysprosium and ACAC and phen). Such device could emit pure linear spectrum (main peak: 480nm and 580nm) of Dy^{3+} under the DC drive of 3V. The quantum efficiency could reach above 1% under the electric current density lower than $200 \mu \text{A}/\text{cm}^2$.

Embodiment 4

The material of the mixed layer is $\text{Er}(\text{DBM})_3\text{phen}$ (the complex of trivalent erbium and DBM and phen), the material of the hole transporting layer is TPD. The proportion of the mixed layer $\text{Er}(\text{DBM})_3\text{phen}:\text{TPD}$ changes from 1:3 to 3:1. Such device could emit pure linear spectrum (main peak: 525nm) of Er^{3+} under the DC drive of 2.5V. The quantum efficiency could reach above 1% under the electric current density lower than $200 \mu \text{A}/\text{cm}^2$.

Embodiment 5

The material of the mixed layer is $\text{Tb}(\text{ACAC})_3\text{phen}$, the material of the hole transporting layer is TPD, and the proportion of the mixed layer $\text{Tb}(\text{ACAC})_3\text{phen}:\text{TPD}$ changes from 1:3 to 3:1, and the material is $\text{Gd}(\text{ACAC})_3\text{phen}$ (the complex of trivalent gadolinium and ACAC and phen). Such device could emit pure linear spectrum (main peak: 545nm) of Tb^{3+} under the DC drive of 3V. The quantum efficiency could reach above 3% under the electric current density lower than $200 \mu \text{A}/\text{cm}^2$.

Embodiment 6

The material of the mixed layer is $\text{Tm}(\text{ACAC})_3\text{phen}$, the material of the hole transporting layer is TPD, the proportion of the mixed layer $\text{Tm}(\text{ACAC})_3\text{phen}:\text{TPD}$ changes from 1:3 to 3:1, and the material of the electron transporting layer is $\text{Gd}(\text{ACAC})_3\text{phen}$ (the complex of trivalent gadolinium and ACAC and phen). Such device could emit pure linear spectrum (main peak: 480nm) of Tm^{3+} under the DC drive of 3V. The quantum efficiency could reach above 3% under the electric current density lower than $200 \mu \text{A}/\text{cm}^2$.

Embodiment 7

The material of the mixed layer is $\text{Nd}(\text{ACAC})_3\text{phen}$, the material of the

hole transporting layer is TPD, the proportion of the mixed layer Nd(ACAC)₃phen:TPD changes from 1:3 to 3:1, and the material of the electron transporting layer is Nd (DBM)₃phen. Such device could emit pure linear spectrum (main peak: 900nm and 980nm) of Nd³⁺ under the DC drive of 6V, and infrared light is emitted.

What is claimed is:

1. An organic electroluminescent device of lanthanide series metal complex with high efficiency and high color purity, whose structure includes a metal electrode plate (1), an electron transporting layer (2), a hole transporting layer (4), an ITO conductive film (5), a transparent glass substrate (6), characterized in that the emission layer is a mixed layer (3), which mixed layer (3) is disposed between the hole transporting layer and the electron transporting layer, and the material of the mixed layer is a mixture of trivalent lanthanide series metal ion complex and the material of the hole transporting layer, and the mix proportion decreases gradually from the hole transporting layer to the electron transporting layer, the material of the electron transporting layer could be either the trivalent lanthanide series metal ion complex InC, or the complex of the non-luminous L lanthanide series metal and C.

2. The organic electroluminescent device of lanthanide series metal complex with high efficiency, high color purity and long life-span according to claim 1, characterized in that in the mixed layer (3), the mix proportion of the trivalent lanthanide series metal ion complex and the material of the hole transporting layer changes gradually from the hole transporting layer from HTL:InC=6:1~3:1 to HTL:LnC=1:6~1:3.

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